FORM PTO-1390 (Modified) (REV 11-2000) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER	U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR
213342US0XPCT	09/926295

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/EP00/02013	8 March 2000	22 April 1999

TITLE OF INVENTION
PROCESS FOR PRODUCING CYANURIC CHLORIDE

APPLICANT(S) FOR DO/EO/US
BOERNER Walter et al.

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. ☒ This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND** or **SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☒ This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include itens (5), (6), (9) and (24) indicated below.
4. ☒ The US has been elected by the expiration of 19 months from the priority date (Article 31).
5. ☒ A copy of the International Application as filed (35 U.S.C. 371 (c) (2))
   a. ☐ is attached hereto (required only if not communicated by the International Bureau).
   b. ☒ has been communicated by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
6. ☒ An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).
   a. ☒ is attached hereto.
   b. ☐ has been previously submitted under 35 U.S.C. 154(d)(4).
7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371 (c)(3))
   a. ☐ are attached hereto (required only if not communicated by the International Bureau).
   b. ☐ have been communicated by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☒ have not been made and will not be made.
8. ☐ An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371 (c)(4)).
10. ☐ An English language translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371 (c)(5)).
11. ☐ A copy of the International Preliminary Examination Report (PCT/IPEA/409).
12. ☒ A copy of the International Search Report (PCT/ISA/210).

Items 13 to 20 below concern document(s) or information included:

13. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
14. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
15. ☒ A **FIRST** preliminary amendment.
16. ☐ A **SECOND** or **SUBSEQUENT** preliminary amendment.
17. ☐ A substitute specification.
18. ☐ A change of power of attorney and/or address letter.
19. ☐ A computer-readable form of the sequence listing in accordance with PCT Rule 13ter.2 and 35 U.S.C. 1.821 - 1.825.
20. ☐ A second copy of the published international application under 35 U.S.C. 154(d)(4).
21. ☐ A second copy of the English language translation of the international application under 35 U.S.C. 154(d)(4).
22. ☐ Certificate of Mailing by Express Mail
23. ☒ Other items or information:

Request for Consideration of Documents Cited in International Search Report
Notice of Priority
Drawings (1 Sheet)

U.S. APPLICATION NO. (IF KNOWN, SEE 37 CFR)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/926295	PCT/EP00/02013	213342US0XPCT

24. The following fees are submitted:.	CALCULATIONS PTO USE ONLY	
BASIC NATIONAL FEE (37 CFR 1.492 (a) (1) - (5)) :		
☐ Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO **$1040.00**		
☒ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO **$890.00**		
☐ International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO **$740.00**		
☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO but all claims did not satisfy provisions of PCT Article 33(1)-(4) **$710.00**		
☐ International preliminary examination fee (37 CFR 1.482) paid to USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) **$100.00**		
ENTER APPROPRIATE BASIC FEE AMOUNT =	**$890.00**	
Surcharge of **$130.00** for furnishing the oath or declaration later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (e)).	**$0.00**	

CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total claims	5 - 20 =	0	x $18.00	**$0.00**	
Independent claims	1 - 3 =	0	x $84.00	**$0.00**	
Multiple Dependent Claims **(check if applicable)**.			☐	**$0.00**	
TOTAL OF ABOVE CALCULATIONS =				**$890.00**	
☐ Applicant claims small entity status. See 37 CFR 1.27). The fees indicated above are reduced by 1/2.				**$0.00**	
SUBTOTAL =				**$890.00**	
Processing fee of **$130.00** for furnishing the English translation later than ☐ 20 ☐ 30 months from the earliest claimed priority date (37 CFR 1.492 (f)). +				**$0.00**	
TOTAL NATIONAL FEE =				**$890.00**	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31) **(check if applicable).** ☐				**$0.00**	
TOTAL FEES ENCLOSED =				**$890.00**	
				Amount to be: refunded	$
				charged	$

a. ☒ A check in the amount of **$890.00** to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. **15-0030** A duplicate copy of this sheet is enclosed.

d. ☐ Fees are to be charged to a credit card. **WARNING:** Information on this form may become public. **Credit card information should not be included on this form.** Provide credit card information and authorization on PTO-2038.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SEND ALL CORRESPONDENCE TO:

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NAME

24,618
REGISTRATION NUMBER

Oct. 9 2001
DATE

DOCKET NO. 213342US0XPCT

IN RE APPLICATION OF: BOERNER Walter et al.

SERIAL NO.: New U.S. PCT Application (Based on PCT/EP00/02013)

FILED: HEREWITH

FOR: PROCESS FOR PRODUCING CYANURIC CHLORIDE

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

Sir:

Transmitted herewith is an amendment in the above-identified application.

☐ No additional fee is required.

☐ Small entity status of this application under 37 C.F.R. §1.9 and §1.27 has been established by a verified statement previously submitted.

☐ Small entity status of this application under 37 C.F.R. §1.9 and §1.27 has been established by a verified statement submitted herewith.

☒ Additional documents filed herewith: PCT Transmittal Letter/Notice of Priority
English Translation of Specification/Request for Consideration/Declaration/Drawings (1 sheet)
Preliminary Amendment/International Search Report/Check for $890.00

The fee has been calculated as shown below.

	(Col. 1)		(Col. 2)	(Col. 3)	SMALL ENTITY		OTHER THAN A SMALL ENTITY	
	CLAIMS REMAINING AFTER		HIGHEST NUMBER PREVIOUSLY PAID FOR	PRESENT EXTRA	RATE	ADDITIONAL FEE	RATE	ADDITIONAL FEE
TOTAL	* 5	MINUS	** 30	= 0	X9 =	$	X18 =	$ .00
INDEP	* 1	MINUS	*** 3	= 0	X40 =	$	X80 =	$ .00
☐	FIRST PRESENTATION OF MULTIPLE DEPENDENT CLAIM				+135=	$	+270=	$
					TOTAL	$	TOTAL	$ .00

___ A check in the amount of $_____ is attached.

XX Please charge any additional fees for the papers being filed herewith and for which no check is enclosed herewith, or credit any overpayment to deposit Account No. 15-0030. A duplicate copy of this sheet is enclosed.

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22850

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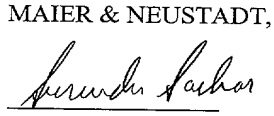

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*If the entry in Column 2 is less than the entry in Column 1 write "0" in Column 3.
**If the "Highest Number Previously paid for" IN THIS SPACE is less than 20 write "20" in this space.
***If the "Highest Number Previously paid for" IN THIS SPACE is less than 3 write "3" in this space.

7/93

213342US-0X PCT

IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :

WALTER BOERNER ET AL : ATTN: APPLICATION DIVISION

SERIAL NO: NEW US PCT APPLN
(Based on PCT/EP00/02013) :

FILED: HEREWITH :

FOR: PROCESS FOR PRODUCING CYANURIC CHLORIDE :

PRELIMINARY AMENDMENT

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, D.C. 20231

SIR:

Prior to examination on the merits, please amend the above-identified application as follows:

IN THE SPECIFICATION

Please replace the title on page 1, line 1 with the following:

--METHOD OF PRODUCING CYANURIC CHLORIDE--.

IN THE CLAIMS

Please amend the claims as shown on the marked-up copy following this amendment to read as follows:

1. (Amended) Process for producing cyanuric chloride, comprising trimerisation of cyanogen chloride in the presence of a washed activated carbon having a BET surface area of

at least 1000 m^2/g and an Fe content of less than 0.15 wt.%, calculated as Fe_2O_3 at a temperature of at least 250 °C,

wherein an activated carbon having an effective pore volume V_{eff} of equal to or greater than 0.17 ml/g is used, V_{eff} is obtained from pores having a pore diameter in the range of 0.5 to 7 nm.

2. (Amended) Process according to claim 1,

wherein the effective pore volume V_{eff} of the activated carbon is calculated from the sum $V_{eff} = 0.25V_{micro} + 0.5V_{meso}$, V_{micro} represents pores having a diameter of less than 2 nm and V_{meso} represents pores having a diameter of 2 to 30 nm.

3. (Amended) Process according to claim 1 wherein

V_{eff} of the activated carbon used is at least 0.2 ml/g.

4. (Amended) Process according to claim 1, wherein

the activated carbon has a bulk density of equal to or less than 420 g/l.

5. (Amended) Process according to claim 1, wherein

the activated carbon has a BET surface area of at least 1200 m^2/g and V_{eff} is at least 0.2 ml/g.

REMARKS

Claims 1-5 are active in the present application. The claims have been amended to remove multiple dependencies and for clarity. No new matter is added. An action on the merits and allowance of claims is solicited.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND,
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213342US-0X PCT

Marked-Up Copy
Serial No:

Amendment Filed on:
10-9-01

IN THE SPECIFICATION

Please replace the title on page 1, with the following:

[OLD TITLE] NEW TITLE

IN THE CLAIMS

Please amend the claims as follows:

--1. (Amended) Process for producing cyanuric chloride, comprising trimerisation of cyanogen chloride in the presence of a washed activated carbon having a BET surface area of at least 1000 m^2/g and an Fe content of less than 0.15 wt.%, [(]calculated as Fe_2O_3[)] at a temperature of at least 250 °C,

[characterised in that] wherein an activated carbon having an effective pore volume V_{eff} of equal to or greater than 0.17 ml/g is used, V_{eff} [being] is obtained from pores having a pore diameter in the range of 0.5 to 7 nm.

2. (Amended) Process according to claim 1,

[characterised in that an activated carbon is used, whose] wherein the effective pore volume V_{eff} of the activated carbon is [formed] calculated from the sum [$V_{eff} = 0.25 \cdot V_{micro} +$

0.5 V_{meso}] $V_{eff} = 0.25V_{micro} + 0.5V_{meso}$, V_{micro} [comprising] represents pores having a diameter of less than 2 nm and V_{meso} [comprising] represents pores having a diameter of 2 to 30 nm.

3. (Amended) Process according to claim 1 [or 2, characterised in that] wherein

V_{eff} of the activated carbon used is at least 0.2 ml/g.

4. (Amended) Process according to [one of claims 1 to 3, characterised in that] claim 1, wherein

the activated carbon [to be used] has a bulk density of equal to or less than 420 g/l.

5. (Amended) Process according to [one of claims 1 to 4, characterised in that] claim 1, wherein

the activated carbon [to be used] has a BET surface area of at least 1200 m^2/g and V_{eff} is at least 0.2 ml/g.--

09/926295
PCT/EP00/02013

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410 Rec'd PCT/PTO 09 OCT 2001

Process for producing cyanuric chloride

Description

This invention relates to a process for producing cyanuric chloride by trimerisation of cyanogen chloride at a temperature of above 200 °C on an activated carbon catalyst. The process according to the invention also results in a decreased specific catalyst consumption.

Cyanuric chloride is produced on a large scale by chlorination of hydrogen cyanide with the formation of cyanogen chloride and trimerisation of the cyanogen chloride to form cyanuric chloride - see Ullmann's Encyclopedia of Industrial Chemistry Vol. A8, 5th ed. (1987), 196-197. The trimerisation is carried out in the vapour phase at a temperature of above 200 °C, in particular in the range of about 300 to 450 °C, on an activated carbon catalyst. During continuous operation, a temperature profile develops along the longitudinal axis of the reactor owing to the exothermicity of the trimerisation reaction; this results in the formation of a so-called hot-spot, the temperature maximum of which depends on the flow rate and rises with increasing flow rate. It is known that the deactivation of the activated carbon catalyst is influenced by the operating conditions, the flow rate and the quality of the activated carbon. The deactivation becomes apparent from the movement of the reaction zone, and with it the temperature maximum, along the longitudinal axis of the catalyst.

Owing to its becoming deactivated, the catalyst has to be exchanged periodically or otherwise activated. The economic efficiency of the cyanuric chloride process depends considerably on the service life of the catalyst, as not only the cost of the catalyst but also the cost of a plant standstill have to be taken into account. Moreover, with increasing deactivation of the catalyst, secondary products

such as, for example, cyameluric chloride, are increasingly discharged and hence necessitate increased expenditure on the purification of the cyanuric chloride.

In view of the problems demonstrated, the experts have for a long time been interested in finding activated carbon catalysts which have an increased service life and/or in varying the operating conditions in such a way that the service life can be increased.

Accordingly, US Patent 3,312,697 discloses a process for producing cyanuric chloride using an activated carbon catalyst having a specific surface of above 1000 m^2/g, in which the activated carbon catalyst was activated by a treatment with acids and/or alkalies and a downstream washing with water. As a result of the above-mentioned treatment, inorganic constituents such as oxides, hydroxides and salts of metals such as Li, Mg, Ce, Ti, V, Mn, Fe, Ni, Pt, Cu, Zn, Cd, Sn, Pb and Bi, which diminish the service life of the catalyst, are dissolved out of the activated carbon. The service life of the catalyst is further increased in this process by the addition of 0.5 to 10 wt.% chlorine and/or phosgene to the cyanogen chloride.

In the process according to US Patent 3,707,544, the service life is increased by mixing the trimerisation reactor with a mixture of an activated carbon and a solid diluent having little or no catalytic activity. The disadvantage of this process is that the space-time yield is diminished and the expense of disposing of the deactivated catalyst is increased, above all if the diluent is a non-combustible material.

In the process described in US Patent 3,867,382, an untreated activated carbon produced from coconut shells is used instead of an acid-washed activated carbon. This activated carbon has an internal surface area of 1200 to 1500 m^2/g, a micropore volume of at least 0.7 cm^3/g and an ash content of below 4 wt.%. Owing to the vegetable origin

of the raw material used for this activated carbon, it has a low content of heavy metals and an acid wash is rendered unnecessary. It cannot be inferred from this document how the micropores are defined, i. e. whether they comprise all the internal pores, or micropores having precisely defined limiting values for the pore diameters. A considerable disadvantage of the activated carbon used in the examples is that the bulk density, and hence the quantity required based on the reactor volume, is very high and thus diminishes the economic efficiency.

In J. Beijing Inst. Chem. Technol. 20 (1993) 1, 55-58, E. Wang et al. explain that several factors, namely, the ash content, the iron content, the specific surface and the pore-size distribution, have to be taken into account when selecting the catalysts for the cyanogen chloride trimerisation. The selection of a suitable activated carbon is complicated by the fact that these factors may mutually influence one another. It is to be concluded from this document that it is advantageous to use a carbon which has as high a specific surface as possible and therefore contains numerous small pores. The latter help to enable the reaction to proceed on a relatively large number of active centres. From the diagrams of the pore-size distribution of two different activated carbons, it is suggested that the pores should have a diameter in particular of less than 2 nm. However, no information can be drawn from the document as to how the individual factors influence the service life of the catalyst in a production plant designed for continuous operation.

Accordingly, the object of the present invention is to demonstrate an improved process for producing cyanuric chloride by trimerisation of cyanogen chloride, the improvement consisting in a decreased specific catalyst consumption. A further object is to demonstrate the criteria whereby the person skilled in the art can select an activated carbon catalyst having an extended service

life for this type of reaction. Other objects can be inferred from the following description of the process according to the invention.

A process for producing cyanuric chloride has been found, comprising trimerisation of cyanogen chloride in the presence of a washed activated carbon having a BET surface area of at least 1000 m^2/g and an Fe content (calculated as Fe_2O_3) of less than 0.15 wt.% at a temperature of at least 250 °C, which is characterised in that an activated carbon having an effective pore volume V_{eff} of equal to or greater than 0.17 ml/g is used, V_{eff} being obtained from pores having a pore diameter in the range of 0.5 to 7 nm. The subclaims are directed towards preferred embodiments of the process.

It was found that the trimerisation of cyanogen chloride proceeds satisfactorily only in those pores having pore diameters in the range of 0.5 to 7 nm, in particular 0.5 to 5 nm; the pore volume of these pores are to be at least 0.17 ml/g. Although the pore distribution of activated carbons can differ very widely depending upon the conditions of their production, the effective pore volumes V_{eff} necessary for the reaction can be defined from the sum of a volume increment for the micropores having a pore diameter of < 2 nm and a volume increment of the mesopores having a pore diameter of 2 to 30 nm. The effective pore volume accordingly can be represented as a linear function: $V_{eff} = a \cdot V_{micro} + b \cdot V_{meso}$. It was also found that the function
$V_{eff} = 0.25 \cdot 0.50\ V_{micro} + V_{meso}$ is a suitable selection criterion for an effective activated carbon having a long service life. The volumes of the micro- and mesopores are determined as follows:-

The micropore volume is determined from the nitrogen adsorption isotherm at the temperature of liquid nitrogen by comparison with a standard isotherm using the t-plot

process of De Boer (cf. De Boer et al. in J. of Colloid and Interface Science 21, 405-44 (1966)) in accordance with DIN 66135, Part 2 (Version of April 1998).

The mesopore volume and the pore distribution are determined from the nitrogen desorption isotherm of Barett, Joyner and Halenda in accordance with DIN 66134 (February 1998). Prior to the measurement, the sample used for the determination of V_{micro} and V_{meso} is treated for 1 h at 200 °C under vacuum (less than 1.3 Pa). The measurement is carried out, for example, in an "ASAP 2400" instrument manufactured by the firm of Micromeritics, Norcross, Ga. (US). The definition of V_{meso} according to the invention includes only mesopores having a diameter of 2 to 30 nm.

A particularly large increase in the service life of the activated carbon in this type of process is achieved if V_{eff} is at least 0.2 ml/g. From an investigation of numerous different activated carbons, it was found that a maximum value of the effective pore volume defined above corresponds to a minimum value of the specific catalyst consumption. Both extremely mesoporous activated carbons and extremely microporous activated carbons have too low a pore volume in the middle pore range, that is, in the range between 0.5 and 5 nm, so that the specific catalyst consumption is considerably higher than in the catalysts to be used according to the invention.

Another feature of the activated carbons to be used according to the invention is the specific surface(BET surface area), which is at least 1000 m^2/g, preferably at least 1200 m^2/g. A high surface area is consequently advantageous, but is not a criterion which allows a conclusion regarding the service life of the catalyst. Thus, different activated carbons having virtually identical specific surfaces exhibit very large differences in their rates of deactivation.

In view of the negative influence of a high iron content on the activated carbon, the iron content, calculated as Fe_2O_3, should be below 0.15 wt.% and preferably around or below 0.1 wt.%. Although an unwashed activated carbon is also catalytically active, in the process according to the invention a washed, in particular an acid-washed, activated carbon is used, because washing is on the one hand a possible way of decreasing the content of iron and of the other heavy metals and hence of minimising the formation of secondary products and, on the other hand, it increases the pore volume, which is important for the reaction. With regard to the minimisation of the specific catalyst consumption, it is moreover advantageous to use a carbon having a bulk density of equal to or less than 420 g/l. Where the activity of the activated carbon catalyst is adequate and the effective pore volume is > 0.17 ml/g, preferably equal or > 0.20 ml/g, it is advantageous that the bulk density be as low as possible. In such cases it is advisable to use an activated carbon having a bulk density of equal to or < 420 g/l, preferably < 390 g/cm^3. Figure 1, which summarises the results of numerous investigations - see Examples - clearly shows the unforeseen extent to which the specific catalyst consumption a (kg catalyst per t of unreacted cyanogen chloride) is dependent on the effective pore volume defined according to the invention when a washed activated carbon having a BET surface area of at least 1000 m^2/g and an Fe content of less than 0.15 wt.% (calculated as Fe_2O_3) is used. The specific catalyst consumption is low, in particular when both the rate of deactivation (the method of determination may be found in the Examples) and at the same time the bulk density of the catalyst are as low as possible.

Examples

The investigations to determine the specific catalyst consumption in the reaction zone during the trimerisation of cyanogen chloride to form cyanuric chloride were carried out in a tubular reactor filled with the activated carbon catalyst being examined. The tubular reactor was cooled by means of a heat-transfer medium; the temperature of the coolant was maintained at 280 °C. The test reactor was connected parallel to an operating reactor. The gaseous cyanuric chloride formed was condensed after having left the reactor and the liquid product was converted into the solid aggregate state by being sprayed into cooled chambers.

The ratio of the length of the reactor to the cross-section of the reactor was 39. During continuous operation, a temperature profile developed along the longitudinal axis of the reactor. This profile comprises a heating zone, a reaction zone and a cooling zone. The maximum of the reaction zone, the temperature of which rises with increasing flow rate, moves forward in the direction of the flow, with increasing deactivation of the catalyst. The rate of deactivation (u_{deact}) was determined by constructing time-dependent temperature profiles from temperature-measuring points arranged along the reactor.

Figure 2 shows that with increasing operating time, the hot-spot of the reaction zone moves through the complete set of measuring points arranged one behind the other. The actual determination of the rate of deactivation was commenced by a so-called preliminary deactivation of the catalyst - at that time, the "hot-spot" developed near to the inlet to the reactor. The preliminary deactivation of the catalyst lasts for about 12 hours at a flow rate of cyanogen chloride of 1.1 kg per hour. Figure 2 shows a typical progression of the deactivation. The rate of deactivation in cm/t ClCN can be determined from the

distance of the temperature-measuring points and the average quantity of cyanogen chloride (measured from maximum to maximum). The specific catalyst consumption in the reaction zone can be determined from the rate of deactivation ($v_{deact.}$), the reactor geometry (cross-sectional area F) and the bulk density ρ, in accordance with the following equation:

$$a\left[\frac{kg\ cat.}{t\ ClCN}\right] = u_{Deact}\left[\frac{cm}{t\ ClCN}\right] \cdot F\ [cm^2] \cdot \rho\left[\frac{kg}{m^3}\right]$$

Table 1: Activated carbon catalysts used

Catalyst (No.)	Raw material	Wash	Ash content (wt.%)	Fe content (as Fe_2O_3) (wt.%)	Bulk density (g/l)	BET (m^2/g)	Pore volumes (cm^3/g)		
							V_{micro}	V_{meso}	V_{eff} *)
C1	Peat	+	1.67	0.00	403	1016	0.38	0.18	0.185
C2	Peat	+	2.45	0.07	346	1453	0.63	0.11	0.213
C3	Hard coal	+	2.24	0.03	410	1217	0.51	0.17	0.212
C4	Wood	+	2.18	0.28	375	1523	0.64	0.09	0.205
C5	Pine wood	-	8.01	0.16	406	1290	0.58	0.11	0.200
C6	Coconut	+	0.42	0.00	373	1459	0.59	0.04	0.157
C7	Peat	+	2.46	0.07	434	1213	0.50	0.08	0.165
C8	Coconut	+	1.66	0.01	430	1110	0.45	0.07	0.147

*) $V_{eff} = 0.25\ V_{micro} + 0.5\ V_{meso}$

Table 2 shows the rate of deactivation u and the specific catalyst consumption a in the reaction zone using the activated carbons given in Table 1, the flow rate of ClCN being 4.4 kg per hour in all the tests.

Table 2: Rate of deactivation V and specific catalyst consumption a in the reaction zone

Catalyst No.	u (cm/t ClCN)	a kg cat./t ClCN
C1	29	1.05
C2	21	0.65
C3	25	0.92
C4 *)	35	1.18
C5 *)	40	1.46
C6 *)	35	1.18
C7 *)	28	1.09

Temperature of the heat-transfer medium: 280 °C
*) activated carbon catalyst not according to the invention

The tests show that the specific catalyst consumption in the reaction zone depends considerably on the effective pore volume and the bulk density of the catalyst. As a result of a decreased consumption of catalyst, not only is the cost of the catalyst decreased, but at the same time the availability of the plant is increased owing to decreased standstill times and the economic efficiency of the process is thereby likewise increased.

Claims

1. Process for producing cyanuric chloride, comprising trimerisation of cyanogen chloride in the presence of a washed activated carbon having a BET surface area of at least 1000 m^2/g and an Fe content of less than 0.15 wt.%, (calculated as Fe_2O_3) at a temperature of at least 250 °C,
characterised in that
an activated carbon having an effective pore volume V_{eff} of equal to or greater than 0.17 ml/g is used, V_{eff} being obtained from pores having a pore diameter in the range of 0.5 to 7 nm.

2. Process according to claim 1,
characterised in that
an activated carbon is used, whose effective pore volume V_{eff} is formed from the sum $V_{eff} = 0.25 \cdot V_{micro} + 0.5\ V_{meso}$, V_{micro} comprising pores having a diameter of less than 2 nm and V_{meso} comprising pores having a diameter of 2 to 30 nm.

3. Process according to claim 1 or 2,
characterised in that
V_{eff} of the activated carbon used is at least 0.2 ml/g.

4. Process according to one of claims 1 to 3,
characterised in that
the activated carbon to be used has a bulk density of equal to or less than 420 g/l.

5. Process according to one of claims 1 to 4,
characterised in that
the activated carbon to be used has a BET surface area of at least 1200 m^2/g and V_{eff} is at least 0.2 ml/g.

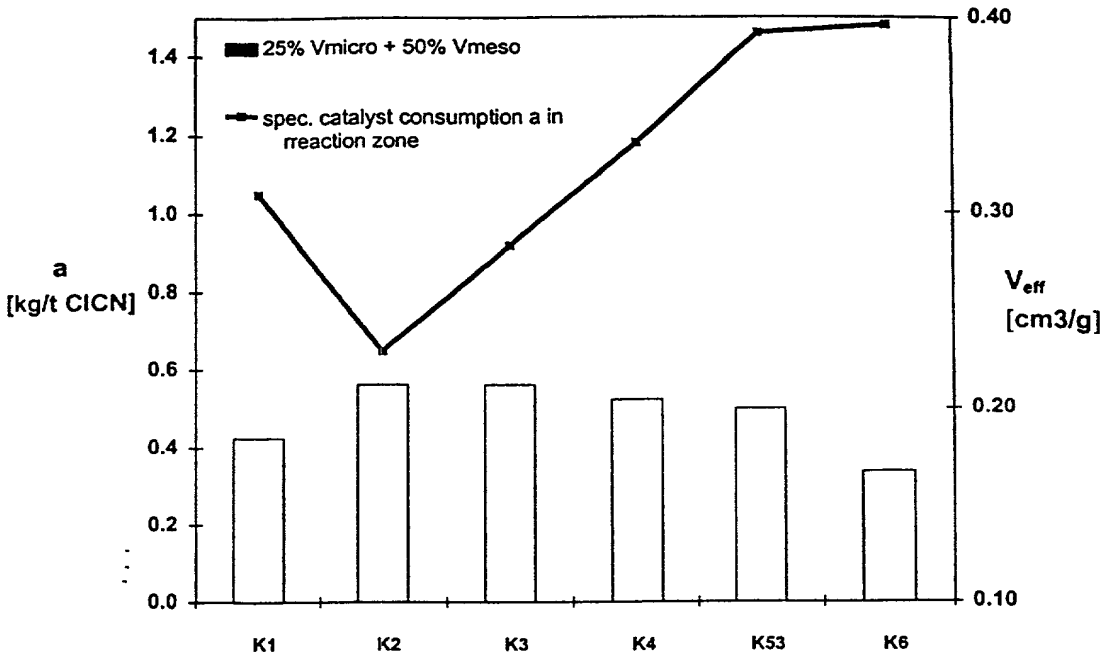

Fig. 1: Specific catalyst consumption a in relation to the effective pore volume

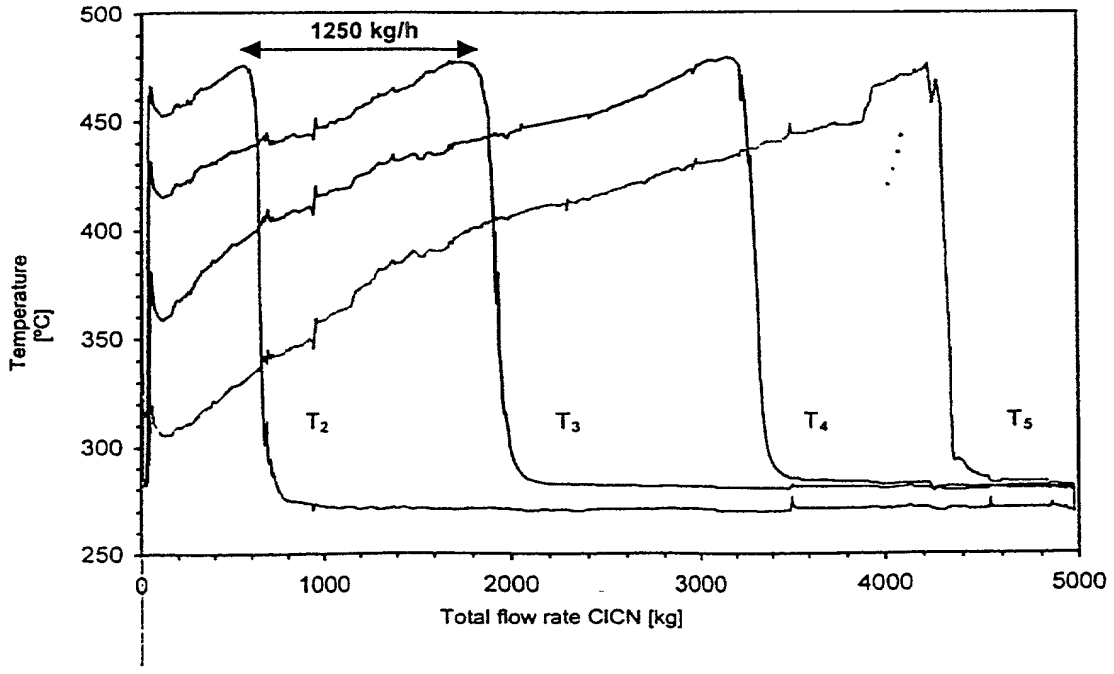

Fig. 2: Movement of the hot spot through the reactor

Declaration and Power of Attorney for Patent Application
Erklärung für Patentanmeldungen mit Vollmacht
German Language Declaration

Als nachstehend benannter Erfinder erkläre ich hiermit an Eides Statt:

daß mein Wohnsitz, meine Postanschrift und meine Staatsangehörigkeit den im nachstehenden nach meinem Namen aufgeführten Angaben entsprechen, daß ich nach bestem Wissen der ursprüngliche, erste und alleinige Erfinder (falls nachstehend nur ein Name angegeben ist) oder ein ursprünglicher, erster und Miterfinder (falls nachstehend mehrere Namen aufgeführt sind) des Gegenstandes bin, für den dieser Antrag gestellt wird und für den ein Patent für die Erfindung mit folgendem Titel beantragt wird:

_____

_____

_____

deren Beschreibung:

☐ ist beigefügt

☐ wurde angemeldet am _____

unter der US-Anmeldenummer oder unter der Internationalen Anmeldenummer im Rahmen des Vertratgs über die Zusammenarbeit auf dem Gebiet des Patentwesens (PCT)

_____ und am

_____ abgeändert (falls zutreffend).

As a below named inventor, I hereby declare that:

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METHOD OF PRODUCING CYANURIC CHLORIDE (as amended)

the specification of which:

☐ is attached hereto.

☒ was filed on 8 March 2000

as United States Application Number or PCT International Application Number

PCT/EP00/02013 and was amended on

_____ (if applicable).

Ich bestätige hiermit, daß ich den Inhalt der oben angegebenen Patentanmeldung, einschließlich der Ansprüche, die eventuell durch einen oben erwähnten Zusatzantrag abgeändert wurde, durchgesehen und verstanden habe.

Ich erkenne meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Titel 37, Code of Federal Regulations, § 1.56 von Belang sind.

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

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Prior foreign application(s)
(Frühere auslandische Anmeldungen)

Priority claimed
Priorität beansprucht

(Number) (Nummer)	(Country) (Land)	(Day/Month/Year Filed) (Tag/Monat/Jahr der Anmeldung)	Yes Ja	No Nein
199 18 245.0	GERMANY	22 April 1999	☒	☐
			☐	☐

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PCT/EP00/02013	8 March 2000	

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

German Language Declaration

VERTRETUNGSVOLLMACHT. Als benannter Erfinder beauftrage ich hiermit den (die) nachstehend aufgeführten Patentanwalt (Patentanwälte) und/oder Vertreter mit der Verfolgung der vorliegenden Patentanmeldung sowie mit der Abwicklung aller damit verbundenen Angelegenheiten vor dem US-Patent- und Markenamt: *(Name(n) und Registrationsnummer(n) auflisten)*

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith: *(list name and registration number)*

022850

Postanschrift:

Send Correspondence to:

022850

Telefonische Auskünfte.
(Name und Telefonnummer)

Direct Telephone calls to: *(name and telephone number)*

(703) 413-3000

Vor- und Zuname des einzigen oder ersten Erfinders	Full name of sole or first inventor Walter BOERNER
Unterschrift des Erfinders Datum	Inventor's signature ✓ Date ✓
Wohnsitz	Residence Lessingstrasse 12, D-63579 Freigericht, Germany
Staatsangehörigkeit	Citizenship Germany
Postanschrift	Post Office Address same as above
Vor- und Zuname des zweiten Miterfinders (falls zutreffend)	Full name of second joint inventor, if any Ralph MARQUARDT
Unterschrift des zweiten Erfinders Datum	Second inventor's signature ✓ Date ✓
Wohnsitz	Residence Schwarzburgstrasse 22, D-60318, Frankfurt am Main, Germany
Staatsangehörigkeit	Citizenship Germany
Postanschrift	Post Office Address same as above

(Im Falle dritter und weiterer Miterfinder sind die entsprechenden Informationen und Unterschriften hinzuzufugen)

(Supply similar information and signature for third and subsequent joint inventors)

German Language Declaration

Vor- und Zuname des dritten Miterfinders (falls Zutreffend)	Full name of third joint inventor, if any Stephanie SCHAUHOFF
Unterschrift des dritten Erfinders Datum	Third inventor's signature ✓ Date ✓
Wohnsitz	Residence Bergerstrasse 152, D-60385 Frankfurt am Main, Germany
Staatsangehörigkeit	Citizenship Germany
Postanschrift	Post Office Address same as above
Vor- und Zuname des vierten Miterfinders (falls Zutreffend)	Full name of fourth joint inventor, if any Christine SCHICK
Unterschrift des vierten Erfinders Datum	Fourth inventor's signature ✓ Date ✓
Wohnsitz	Residence Bettinastrasse 66, D-63067 Offenbach, Germany
Staatsangehorigkeit	Citizenship Germany
Postanschrift	Post Office Address same as above
Vor- und Zuname des fünften Miterfinders (falls Zutreffend) 5-CJ	Full name of fifth joint inventor, if any Rudolf VANHEERTUM
Unterschrift des fünften Erfinders Datum	Fifth inventor's signature Rudolf VanHeertum Date 9.07.01
Wohnsitz	Residence Antoinettalei 11, BE-2930 Brasschaat, Belgium BE
Staatsangehörigkeit	Citizenship Belgium
Postanschrift	Post Office Address same as above
Vor- und Zuname des sechsten Miterfinders (falls Zutreffend)	Full name of sixth joint inventor, if any
Unterschrift des sechsten Erfinders Datum	Sixth inventor's signature Date
Wohnsitz	Residence
Staatsangehörigkeit	Citizenship
Postanschrift	Post Office Address

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METHOD OF PRODUCING CYANURIC CHLORIDE (as amended)

deren Beschreibung:

- ☐ ist beigefügt
- ☐ wurde angemeldet am _____

  unter der US-Anmeldenummer oder unter der Internationalen Anmeldenummer im Rahmen des Vertratgs über die Zusammenarbeit auf dem Gebiet des Patentwesens (PCT)

  _____ und am

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- ☐ is attached hereto.
- ☒ was filed on 8 March 2000

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199 18 245.0	GERMANY	22 April 1999	☒ Yes Ja	☐ No Nein
			☐ Yes Ja	☐ No Nein

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PCT/EP00/02013	8 March 2000	

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Vor- und Zuname des einzigen oder ersten Erfinders	Full name of sole or first inventor Walter BOERNER
Unterschrift des Erfinders Datum	Inventor's signature ✓ Date ✓
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Postanschrift	Post Office Address same as above
Vor- und Zuname des zweiten Miterfinders (falls zutreffend) 2-00	Full name of second joint inventor, if any Ralph MAROUARDT
Unterschrift des zweiten Erfinders Datum	Second inventor's signature ✓ Ralph Marquardt ✓ Date 09/16/01
Wohnsitz	Residence Usastvasse 1, D-60388, Frankfurt am Main, Germany
Staatsangehörigkeit	Citizenship Germany DEX
Postanschrift	Post Office Address same as above

(Im Falle-dritter und weiterer Miterfinder sind die entsprechenden Informationen und Unterschriften hinzuzufugen.)

(Supply similar information and signature for third and subsequent joint inventors.)

German Language Declaration

Vor- und Zuname des dritten Miterfinders (falls Zutreffend)	Full name of third joint inventor, if any Stephanie SCHAUHOFF
Unterschrift des dritten Erfinders Datum	Third inventor's signature Date
Wohnsitz	Residence Bergerstrasse 152, D-60385 Frankfurt am Main, Germany
Staatsangehorigkeit	Citizenship Germany
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Unterschrift des vierten Erfinders Datum	Fourth inventor's signature Date
Wohnsitz	Residence Bettinastrasse 66, D-63067 Offenbach, Germany
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Postanschrift	Post Office Address same as above
Vor- und Zuname des fünften Miterfinders (falls Zutreffend)	Full name of fifth joint inventor, if any Rudolf VANHEERTUM
Unterschrift des fünften Erfinders Datum	Fifth inventor's signature Date
Wohnsitz	Residence Antoinettalei 1, BE-2930 Brasschaat, Belgium
Staatsangehörigkeit	Citizenship Belgium
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Vor- und Zuname des sechsten Miterfinders (falls Zutreffend)	Full name of sixth joint inventor, if any
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(Application No.) (Aktenzeichen)	(Filing Date) (Anmeldetag)	(Application No.) (Aktenzeichen)	(Filing Date) (Anmeldetag)

Ich beanspruche hiermit die mir unter Title 35, US-Code, § 120 zustehenden Vorteile aller unten aufgeführten US-Patentanmeldungen bzw. § 365(c) aller PCT internationalen Anmeldungen, welche die Vereinigten Staaten von Amerika benennen, und erkenne, insofern der Gegenstand eines jeden früheren Anspruchs dieser Patentanmeldung nicht in einer US-Patentanmeldung, bzw. PCT internationalen Anmeldung in in einer gemäß dem ersten Absatz von Title 35, US-Code, § 112 vorgeschriebenen Art und Weise offenbart wurde, meine Pflicht zur Offenbarung jeglicher Informationen an, die zur Prüfung der Patentfähigkeit in Einklang mit Title 37, Code of Federal Regulations, § 1.56 von Belang sind und die im Zeitraum zwischen dem Anmeldetag der früheren Patentanmeldung und dem nationalen oder im Rahmen des Vertrags über die Zusammenarbeit auf dem Gebiet des Patentwesen (PCT) gültigen internationalen Anmeldetags bekannt geworden sind

I hereby claim the benefit under Title 35, United States Code, § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of Title 35, United States Code, § 112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

(Application No.) (Aktenzeichen)	(Filing Date) (Anmeldetag)	(Status) (patented, pending, abandoned) (Status) (patentiert, schwebend, aufgegeben)
PCT/EP00/02013	8 March 2000	

Ich erkläre hiermit, daß alle in der vorliegenden Erklärung von mir gemachten Angaben nach bestem Wissen und Gewissen der Wahrheit entsprechen, und ferner daß ich diese eidesstattliche Erklärung in Kenntnis dessen ablege, daß wissentlich und vorsätzlich falsche Angaben oder dergleichen gemäß § 1001, Title 18 des US-Code strafbar sind und mit Geldstrafe und/oder Gefängnis bestraft werden können und daß derartige wissentlich und vorsätzlich falsche Angaben die Rechtswirksamkeit der vorliegenden Patentanmeldung oder eines aufgrund deren erteilten Patentes gefährden können.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

German Language Declaration

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POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith *(list name and registration number)*

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German Language Declaration

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